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Pyrochemical Treatment of Spent Nuclear Fuel

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ABSTRACT: Over the last 10 years, pyrochemical treatment of spent nuclear fuel has progressed from demonstration activities to engineering-scale production operations. As part of the Advanced Fuel Cycle Initiative within the U.S. Department of Energy's Office of Nuclear Energy, Science and Technology, pyrochemical treatment operations are being performed as part of the treatment of fuel from the Experimental Breeder Reactor II at the Idaho National Laboratory. Integral to these treatment operations are research and development activities that are focused on scaling further the technology, developing and implementing process improvements, qualifying the resulting high-level waste forms, and demonstrating the overall pyrochemical fuel cycle.

KEYWORDS: *pyrochemical, pyroprocessing, electrometallurgical, electrorefining*

I. INTRODUCTION

As part of the Advanced Fuel Cycle Initiative (AFCI) within the U.S. Department of Energy's Office of Nuclear Energy, Science and Technology, both aqueous and non-aqueous fuel treatment technologies are being developed. Pyrochemical treatment processes are one type of non-aqueous technology that is being developed and implemented at the engineering scale. Much of the development of this technology is based on treatment of spent Experimental Breeder Reactor II (EBR-II) fuel. Treatment of the EBR-II fuel has been ongoing in the Fuel Conditioning Facility (FCF), located at the Materials and Fuel Complex (MFC) of the Idaho National Laboratory (INL) since 1996.

The AFCI program supports two key aspects of pyrochemical treatment. Most importantly, AFCI focuses on research and development aimed at resolving technical uncertainties, scaling up the technology and expanding the utility of the process to a range of fuel types. Simultaneously, the program supports treatment and qualification of the spent EBR-II fuel for final disposal. EBR-II was a sodium-cooled fast reactor. Its fuel was sodium-bonded metallic fuel. This reactor and fuel type are both considered candidates for Generation IV reactor concepts.

For the purposes of this paper, pyrochemical process operations are divided into two areas, fuel treatment and high-level waste production. The first step in treatment operations is chopping the spent fuel and loading the segments into steel baskets. The steel baskets are transferred into an electrorefiner. In the electrorefiner they serve as an anode. The electrorefiner contains a molten salt medium of LiCl-KCl eutectic and dissolved actinide chlorides, such as UCl_3 and $PuCl_3$.

In the electrorefiners, the spent fuel is electrochemically dissolved from the anode baskets, and an equivalent amount of uranium is deposited on a cathode. The uranium is separated from the bulk of the fission products and transuranics. Most of the fission products (alkali, alkaline earth, rare earth, and halides) and transuranics accumulate in the salt. The bond sodium is neutralized by forming NaCl. The cathode products from electrorefining operations are further processed by distilling adhering salt from the recover uranium.

Pyrochemical treatment of spent nuclear fuel for disposition results in two high-level waste (HLW) forms, the ceramic waste form and the metal waste form. The ceramic waste form, which stabilizes the electrorefiner salts, is a glass-bonded sodalite produced from the thermal conversion of zeolite A. The salts are occluded into the zeolite structure in a heated V-mixer. After the salt is occluded in the V-mixer, the salt–loaded zeolite is mixed with 25% glass frit. This mixture is loaded into a canister and then consolidated into a monolithic waste form in a furnace at 915ºC.

The metal waste form consists of metallic ingots that are used to stabilize noble metal fission products, non-actinide fuel matrix, and cladding materials. Zirconium metal is added to improve performance properties and to produce a lower melting point alloy. The typical composition is stainless steel and 15 weight percent zirconium. It is produced in a casting operation at 1600ºC.

II. Fuel Treatment

Scale-up of pyrochemical treatment operations has been a focus for the last several years. The main goals of scaling the technology are to reduce costs and duration for treating EBR-II fuel and to demonstrate the potential for further application of the technology. To date, electrorefining technologies implemented in a remote hot-cell environment have been scaled by more than three orders of magnitude.

Three different electrorefiners are presently operated in the hot cells at INL. The current capacities in these systems range from 3.5 amps to 2400 amps.

The first of these electrorefiners is called the Hot Fuel Dissolution Apparatus (HFDA) and is installed in the Hot Fuel Examination Facility (HFEF) at MFC. This electrorefiner was placed into operation in the late 1980s. It was used for initial testing of the pyrochemical process with spent fuel at laboratory scale. The current capacity of the HFDA is approximately 3.5 amps. The current efficiency is approximately 50%. This electrorefiner (Figure 1) is still operational. Its maximum batch size is approximately 50 grams.

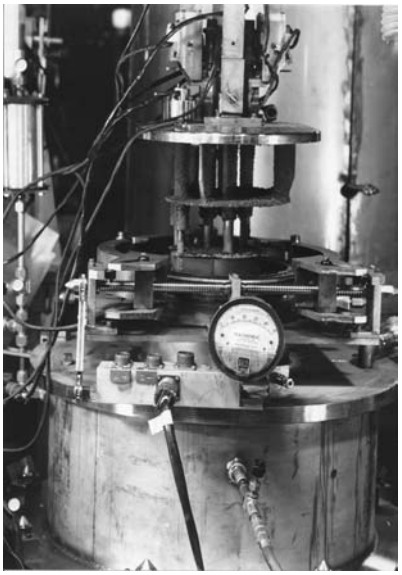

Figure 1

The second electrorefiner became operational in 1995. It is called the Mark IV electrorefiner (the Mark I, II, and II electrorefiners were used for testing with surrogate materials at Argonne National Laboratory in Illinois). The Mark IV electrorefiner has two 300-amp power supplies. Typically, each power supply is only operated at 100 amps. The current efficiency for this electrorefiner is also approximately 50%. The Mark IV electrorefiner batch size using two anode baskets is 24 kg of heavy metal (kghm). The fuel processed in this system is typically highly enriched uranium. This electrorefiner has now been operating for over 10 years in FCF.

The third electrorefiner, the Mark V, was placed into operation in 1998. A picture taken through an FCF hot cell window is provided in Figure 2. The Mark IV and V electrorefiners use identically sized vessels and have the same in-cell footprints. The Mark V electrorefiner has four 600-amp power supplies, and because of improved anode and cathode arrangements, the current efficiency for this system is approximately 80%.

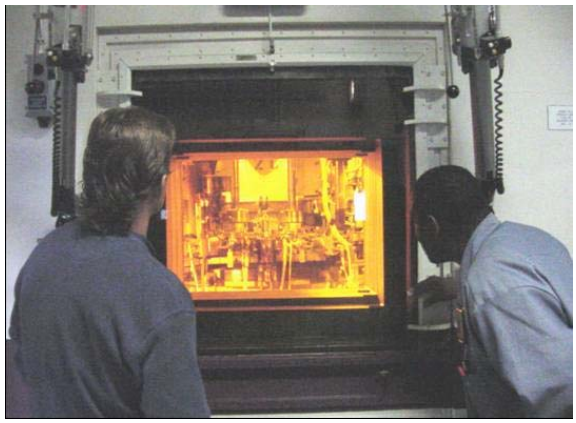

Figure 2

Between the HFDA and Mark V electrorefiner, the potential increase in throughput as controlled by current is greater than a factor of 1000. When comparing the Mark IV and V electrorefiners, the throughput is increased by almost a factor of 20 in identically sized vessels. This scale up was accomplished in three years which included startup testing in the Mark IV electrorefiner and simultaneous development and design of the Mark V electrorefiner.

Establishing performance data has also been a focus of electrorefining work with spent fuel. One key performance aspect is the dissolution of spent fuel. Early tests in the HFDA employed uranium-plutonium based metal fuels. These tests demonstrated the ability to dissolve at least 99.7% of the transuranics and uranium in the spent fuel. The Mark IV and V electrorefiners have been used to just process uranium-based fuels but at much larger batch sizes. In these operations, the ability to dissolve at least 99.7% of the actinides has also been demonstrated. [1]

Tests are also being performed to optimize further the performance of these existing systems and to obtain additional data to support the design of new systems. Current efficiencies in the Mark IV electrorefiner have been recently increased from 50% to 80%. New cathode material studies are ongoing to further improve performance.

The application of this technology to treatment of fuel types other than metallic fuel is also being assessed with spent fuel. Limited process testing with oxide-based fuels was performed in 2003 and 2005. These laboratory-scale tests were completed with 50-gram loadings of irradiated oxide fuel. Initial tests indicated high reduction values (in excess of 98%). Additional laboratory-scale testing is underway.[2]

Limited work on the group recovery of transuranics from pyrochemical processing has also been accomplished. Tests with irradiated materials have been performed at both the laboratory and engineering scale. Laboratory-scale work was performed in the HFDA and engineering-scale work was performed in the Mark V electrorefiner. Separation factor data obtained indicate that the transuranics including plutonium are recovered together along with some of the

lanthanide fission products. The amount of fission products are low enough to not affect fuel performance in fast reactors. Transuranic recoveries at the kilogram scale (Figure 3) have been demonstrated.

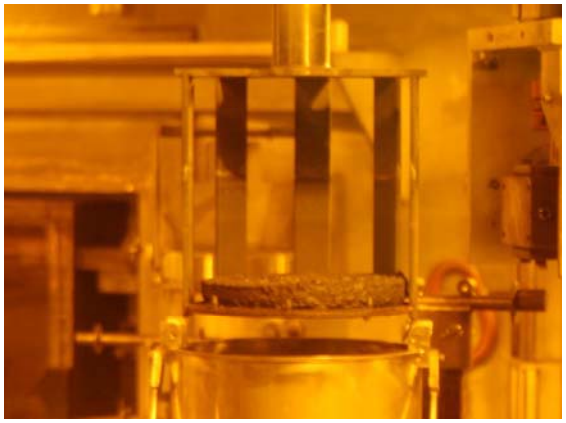

Figure 3

III. High-Level Waste Qualification and Production

Significant effort has been focused on completing qualification of the two high-level waste forms that are generated via pyrochemical fuel treatment. Activities are underway to support both qualification of the waste forms and qualification of the respective production processes. Extensive characterization activities have been performed on the waste forms produced from the treatment of the irradiated EBR-II fuel.

A production furnace for the metal waste form (Figure 4) is presently being readied for installation and operation in the hot cells, while a prototype furnace (Figure 5) is being operated in a glovebox for the development of production process parameters. The production furnace is scheduled for installation in HFEF in 2006. The equivalency of the prototype furnace to the production furnace was demonstrated through a series of instrumented process tests. The prototype furnace is now being used to produce a number of representative waste forms under identical process conditions to demonstrate the reproducibility of the production process.

Figure 4

Figure 5

A consolidation furnace (Figure 6) for the production of full-scale ceramic waste has been procured, installed, and is ready for operation and process testing out of cell. Process testing will first be performed with waste form surrogates before installation in a hot cell. This furnace is capable of producing 400 kg waste forms. Other equipment used for the production of the ceramic waste form, including a large heated V-mixer and a mill/classifier, are already installed and operational in HFEF (Figures 7 and 8).

Figure 6

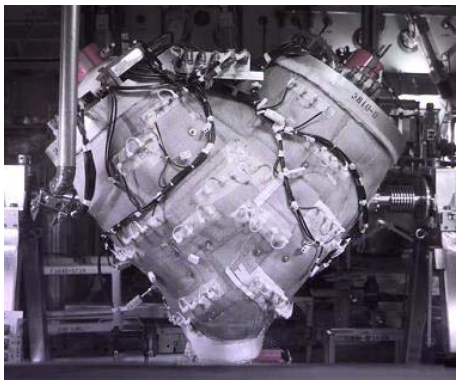

Figure 7

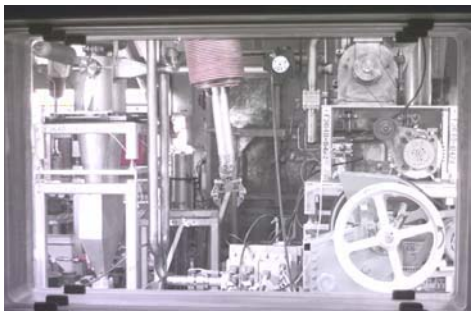

Figure 8

IV. CONCLUSIONS

Over the last 10 years, electrometallurgical treatment has progressed from a demonstration technology to an engineering-scale production process for treating sodium-bonded spent nuclear fuel. As the work progressed to the engineering-scale, key performance data like fuel dissolution efficiencies and current efficiencies have been maintained or even improved compared to laboratory-scale tests. Additionally, under the AFCI Program, key demonstrations have been made in the areas of transuranic recovery at both the laboratory and engineering scales and of reduction of oxide fuels at the laboratory scale. Final disposition of resulting wastes have been an integrated component in the development of these processes. Concurrent with work on fuel treatment processes, HLW form production processes are being designed, tested, and implemented. Overall fuel treatment rates are being maintained while research and development activities are supported to further increase process throughputs and to expand the applicability of pyrochemical processing in the nuclear fuel cycle.

ACKNOWLEDGMENTS

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